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to the
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for research on
BASIC OPTICAL STUDIES OF HIGH T_c OXIDE SUPERCONDUCTORS

AFOSR Agreement *#90–0123*
for the period
January 15, 1990 — January 14, 1993

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Principal Investigator: M.S. Dresselhaus
Co-Principal Investigator: G. Dresselhaus
Room 13-3005
Massachusetts Institute of Technology
Center for Materials Science and Engineering
77 Massachusetts Avenue
Cambridge, MA 02139
Tel. (617) 253-6864

June 15, 1993

Annual Report

Abstract

During the 1992–93 year of the research program on "Basic Optical Studies of High T_c Oxide Superconductors", the effort was focused on a number of projects. The collaborative study of the microwave properties of superconductors with researchers at Rome Labs and the MIT Lincoln Laboratory continued and significant results were obtained on the microwave behavior in the non-linear high rf power regime for $YBa_2Cu_3O_{7-x}$ and this work will be a major part of the Ph.D. thesis of Paul Nguyen. During this year we continued study of the effect of halogen doping on the restoration of superconductivity to YBCO samples previously deoxygenated, and hence rendered non-superconducting. Extensive progress was also made on our new effort in studying superconductivity and the associated phonon spectra and electron-phonon interaction in the alkali metal doped C_{60} materials. We review in this report the progress that was made on each of these projects, referring to the publications that were prepared on each of the topics under investigation.

Microwave Properties of High T_c Superconductors

A main topic under investigation has been the experimental measurement and theoretical modeling of the microwave properties of high temperature superconductors, carried out as a collaborative effort between researchers at MIT (including an MIT Physics graduate student Paul Nguyen, who is working on his Ph.D. thesis), Rome Labs and the MIT Lincoln Laboratory.

The nonlinearity of the surface resistance R_s of $YBa_2Cu_3O_{7-x}$ as a function of the microwave power level is of great importance for passive microwave devices. Since R_s depends upon the microwave current I_{rf} and microwave magnetic field H_{rf}, microwave circuit designs must consider the losses, harmonic generation and intermodulation distortion at the operating power levels. Microwave frequency $1/f$ noise may also be related to nonlinear conduction processes. The dependence of R_s on the microwave magnetic field ($R_s(H_{rf})$) must therefore be measured, since materials with comparable low-field R_s values can differ substantially in their rf field dependencies. To address this issue, we have carrier out the first complete characterization of the nonlinear properties of high-quality $YBa_2Cu_3O_{7-x}$ thin films by measuring $R_s(H_{rf})$ for H_{rf} from 0 to approximately 1 kOe, versus temperature from 4.2K to T_c, and versus frequency from 1.5 GHz to 20 GHz using a stripline resonator. At all frequencies, there is a sharp increase in the nonlinearity at about 60 K. At low fields, flux penetration at weak links dominates the behavior, while at high fields and temperatures, losses

due to pair breaking or flux flow cause the nonlinearities. In this work we have explored the various mechanisms causing the nonlinearities and how the field dependencies are related to film structure and quality. The results have been interpreted in terms of a coupled-grain model which shows good agreement with the experimentally measured $R_s(H_{rf})$. The imaginary part of $Z_s(H_{rf})$, i.e., $X_s(H_{rf})$, however, does not seem to fit the model.

Several publications on this topic have been completed during the grant period January 15, 1992 – January 14, 1993 (#1,2,6,9).

Halogen substitution experiments

The restoration of superconductivity to partially de-oxygenated non-superconducting $YBa_2Cu_3O_{6.2}$ crystals through halogen substitution (#4) has stimulated a large amount of scientific and technological interest in the compound $YBa_2Cu_3O_xX_y$ where X is a halogen. Since bromine addition can be carried out at much lower temperatures ($\sim$ 230°C) than oxygen annealing, the halogenation approach for the restoration of T_c is of technological interest.

To carry out transport measurements including temperature dependent resistivity and magnetoresistance measurements, we have prepared de-oxygenated non-superconducting $YBa_2Cu_3O_{6.2}$ single crystals and we have subsequently doped them with Br. The resulting crystals become superconducting with $T_c \sim 92$ K, $\Delta T_c \sim 1.0$ K as determined by four-point probe resistivity measurements. The normal state resistivity in our best sample decreases linearly with decreasing temperature. We found a large ratio in the resistivity at 300 K of the brominated to the pristine YBCO single crystals (~35:1), suggesting that bromination greatly increases the scattering rate. For these samples we have determined by resistivity measurements the upper critical field $H_{c2}(T)$ parallel and perpendicular to the ab plane. $H_{c2}(0)$ and the corresponding coherence lengths $\xi_{ab}(0)$ and $\xi_c(0)$ were then estimated from the Ginzburg-Landau relations. A comparison with the fully oxygenated YBCO single crystals show that $\xi_{ab}(0)$ remains approximately the same upon bromination, whereas $\xi_c(0)$ decreases by a factor of ~ 3, suggesting that Br never enters the CuO_2 planes. The pinning energy for vortex motion in the ab plane decreases after bromination and this decrease can be attributed to the increased anisotropy. For vortex motion along the $\hat{c}$-axis, however, the pinning energy decreases or increases depending on temperature and field. Compared with the fully oxygenated YBCO single crystals, the critical current density is suppressed by bromination and is strongly dependent on the applied magnetic field. The reduced lower critical field H_{c1} in the brominated YBCO single crystals indicates a reduction in the carrier density (by a factor of between 2 and 3), which is consistent with the observed reduction in the plasma frequency.

A paper based on this work has been prepared and accepted for publication (#4).

Phonons and Electron-Phonon Coupling in C_{60} and Doped C_{60}

The alkali metal doped C_{60} compounds are particularly interesting because of their relatively high values of T_c. Because of the importance of the electron-phonon interaction in superconductors, we have initiated a detailed study of phonons and the electron-phonon interaction in alkali metal doped C_{60}. Our initial work has addressed the phonon spectrum and subsequently the electron-phonon interaction in undoped and doped C_{60} will be studied.

A force constant model has been developed for the vibrational modes in C_{60}, based on bond-stretching and angle-bending interactions (#3). The result of this model are compared with the experimental data available from Raman, infrared, and high resolution electron energy loss spectroscopies, as well as neutron inelastic scattering measurements. Excellent agreement is obtained between calculation and experiment. This force constant model not only provides the mode frequencies but also all the eigenvectors. This model for C_{60} has subsequently been extended to treat C_{70} and alkali metal doped C_{60} and C_{70} (#11,12). These results have been very useful for the interpretation of experimental Raman spectra in C_{70}, M_xC_{60} and M_xC_{70} films (#17). A model for the phonon spectra of carbon nanotubes has also been developed (#18) utilizing the generalized symmetry properties of carbon nanotubes (#15).

An expression for the electron-phonon coupling strength in fullerene nanotubules has been derived within the tight binding model. It is estimated that under ideal circumstances, the electrical conductivity of metallic fullerene nanotubules could be an order of magnitude larger than that of copper. What is of particular interest is the limited availability of scattering states for such a 1D electron system and the special restrictions for scattering of longitudinal modes (intra-branch scattering only) and transverse modes (inter-branch scattering only). This work has been submitted for publication (#16).

The influence of curvature on the electron-phonon coupling in C_{60} doped with alkali metals has been studied within the tight-binding formulation. It is shown that the modulation of the tight binding matrix element due to the displacement of atoms from equilibrium has a value which is essentially the same as in graphite. Nevertheless, the existence of coupling between the electrons and the low-frequency radial phonons in M_3C_{60}, M being an alkali-metal atom, leads to a reasonably large value of λ, the dimensionless electron-phonon coupling parameter. The superconducting transition temperature is then readily calculated and is found to be in the experimentally observed range (#8).

Experimental transport studies of weak localization effects in the normal state of superconducting K_3C_{60} films have been carried out (#10). The femtosecond dynamics of K_3C_{60} and Rb_3C_{60} films have been investigated experimentally and related to that observed in C_{60} films before intercalation, showing a great enhancement in coupling between electrons and phonons in the metallic M_3C_{60} system relative to that in the precursor insulating C_{60} film. These results have been related to interband processes in the respective materials. The relation of these findings to the observation of superconductivity in M_3C_{60} films has also been discussed (#14).

Several review articles relevant to fullerenes were prepared (#5,13).

Invited Talks and Honors

Honors

- Elected Treasurer of the National Academy of Sciences
- Honorary D.Sc., University of Massachusetts, Boston
- Honorary D.Sc., University of Connecticut
- Honorary D.Sc., Princeton University
- Houston Lecturer, Rice University

Research Lectures related to work supported by this grant

- Jan. 16, 1992
  General Motors Research Laboratory
- Jan. 21, 1992
  MIT Club of Boston
- Jan. 28, 1992
  George Washington University and the National Science Foundation
- Feb. 17, 1992
  WPI
- Mar. 3, 1992
  Williams College
- Apr. 30, 1992
  MIT, Math Department Lecture
- May 19, 1992
  Electrochemical Society, Invited talk

- May 21, 1992
  CalTech Physics Colloquium
- May 26, 1992
  AT&T Bell Laboratory
- May 27, 1992
  Princeton University Symposium
- Aug. 3, 1992
  Hokkaido - Hironaka Symposium
- Oct. 1, 1992
  Emery University
- Oct. 14, 1992
  NEC Symposium, Karuizawa, Japan; invited talk
- Oct. 15, 1992
  NEC Symposium, Karuizawa, Japan; Summary talk
- Nov. 29, 1992
  MRS Tutorial on Fullerenes
- Dec. 8, 1992
  University of Maryland
- Jan. 10, 1993
  Houston Lecture, Rice University
- Jan. 12, 1993
  Houston Lecture, Rice University

New Discoveries, Patents or Inventions

None.

Publications

1. C. C. Chin, D. E. Oates, G. Dresselhaus, and M. S. Dresselhaus, Nonlinear electrodynamics of superconducting NbN and Nb thin films at microwave frequencies, Phys. Rev. **B45**, 4788 (1992).
2. C. C. Chin, D. E. Oates, G. Dresselhaus, and M. S. Dresselhaus. Nonlinear microwave electrodynamics of superconducting NbN and Nb thin films, In *Advances in Superconductivity IV*, edited by H. Hayakawa

and N. Koshizuka, page 167, Springer–Verlag, Tokyo, Berlin, New York, 1991. Proceedings of the 4^{th} International Symposium on Superconductivity (ISS'91), October 14-17, 1991, Tokyo.

3. R. A. Jishi, R. M. Mirie, and M. S. Dresselhaus, Force constant model for the vibrational modes in C_{60}, Phys. Rev. **B45**, 13685 (1992).

4. P. P. Nguyen, Z. H. Wang, A. M. Rao, M. S. Dresselhaus, J. S. Moodera, G. Dresselhaus, H. B. Radousky, R. S. Glass, and J. Z. Liu, Transport and Magnetic Properties of $YBa_2Cu_3O_{7-\delta}Br_y$ Single Crystals, Phys. Rev. **B46**, xxx (1993). in press, BV4786.

5. M. S. Dresselhaus, G. Dresselhaus, and P. C. Eklund, in *Encyclopedia of Applied Physics*, page 515, edited by Rita G. Lerner and George L. Trigg (AIP, New York, 1993). Fullerenes, Volume 6.

6. D. E. Oates, P. P. Nguyen, G. Dresselhaus, M. S. Dresselhaus, C. W. Lam, and S. M. Ali, Measurements and Modeling of Linear and Nonlinear Effects in Striplines, J. of Superconductivity **5**, 361 (1992). Conference Proceedings.

7. R. A. Jishi, R. M. Mirie, and M. S. Dresselhaus. A model calculation for the vibrational modes in C_{60}, In *Novel Forms of Carbon: MRS Symposia Proceedings, San Francisco, volume* **270**, edited by C. L. Renschler, J. Pouch, and D. Cox, page 197. Materials Research Society Press, Pittsburgh, PA, 1992.

8. R. A. Jishi and M. S. Dresselhaus. Electron-Phonon Coupling in Superconducting Alkali-Metal Doped Fullerenes, In *Novel Forms of Carbon: MRS Symposia Proceedings, San Francisco, volume* **270**, edited by C. L. Renschler, J. Pouch, and D. Cox, page 161. Materials Research Society Press, Pittsburgh, PA, 1992.

9. D. E. Oates, P. P. Nguyen, G. Dresselhaus, M. S. Dresselhaus, and C. C. Chin, Nonlinear Surface Resistance in $YBa_2Cu_3O_{7-x}$ Thin Films, (1993). Proceedings of the Applied Superconductivity Conference, IEEE Trans. Appl. Superconductivity **3**, 1114.

10. Z. H. Wang, A. W. P. Fung, G. Dresselhaus, M. S. Dresselhaus, K. A. Wang, P. Zhou, and P. C. Eklund, Weak localization and electron-electron interactions in K_3C_{60} films, Phys. Rev. **B47** (1993). in press.

11. R. A. Jishi, R. M. Mirie, M. S. Dresselhaus, G. Dresselhaus, and P. C. Eklund, Force constant model for the vibrational modes in C_{70}, Phys. Rev. **BXX** (1993). accepted, 2/17/93: BB5231.

12. R. A. Jishi, M. S. Dresselhaus, G. Dresselhaus, K. A. Wang, Ping Zhou, A. M. Rao, and P. C. Eklund, Vibrational Mode Frequencies in C_{70}, Chem. Phys. Lett. **206**, 187 (1993).

13. M. S. Dresselhaus, G. Dresselhaus, and P. C. Eklund, Fullerenes, J. Mater. Res. **8** (1993). in press.

14. S. B. Fleischer, E. P. Ippen, G. Dresselhaus, M. S. Dresselhaus, A. M. Rao, P. Zhou, and P. C. Eklund, Femtosecond optical dynamics of C_{60} and M_3C_{60}, Appl. Phys. Lett. **25** (1993). in press.

15. R. A. Jishi, M. S. Dresselhaus, and G. Dresselhaus, Symmetry Properties of Chiral Carbon Nanotubes, Phys. Rev. **BXX** (1993). PRB Rapid Comm. accepted 4/9/93: BBS510.

16. R. A. Jishi and M. S. Dresselhaus, Electron-Phonon Coupling and the Electrical Conductivity of Fullerene Nanotubules, Phys. Rev. (1993). submitted 2/22/93: BB5261.

17. K. A. Wang, Ping Zhou, A. M. Rao, P. C. Eklund, M. S. Dresselhaus, and R. A. Jishi, Intramolecular Vibrational Mode Softening in Alkali-Metal Saturated C_{70} Films, Phys. Rev. (1993). accepted.

18. R. A. Jishi, L. Venkataraman, M. S. Dresselhaus, and G. Dresselhaus, Phonon Modes in Carbon Nanotubules, Chem. Phys. Lett. (1993). accepted, 4/21/93.